

Argonne National Laboratory

**SOME ASPECTS OF THE APPLICATION
OF NUCLEAR ENERGY TO SMALL
PORTABLE AND AUTOMOTIVE
POWER SUPPLIES**

by

D. R. MacFarlane

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SOME ASPECTS OF THE APPLICATION OF NUCLEAR ENERGY
TO SMALL PORTABLE AND AUTOMOTIVE POWER SUPPLIES

by

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ABSTRACT

The problem of applying nuclear energy to segments of the United States economy other than central station electric power, specifically to small portable power supplies used for transportation purposes, is treated. A survey of various energy conversion devices suitable for use in mobile power units is presented. Included also is a comparison of a number of energy source-converter combinations for application as portable power units. It is concluded that storage of nuclear energy in chemical fuels is the only feasible way of approaching the performance of present day, internal combustion engines with hydrocarbon fuels in a future system dependent totally on nuclear energy. A preliminary survey of likely chemical fuels for such a nuclear-chemical energy distribution system is presented. Materials which approach hydrocarbons on an energy density basis are light elements in the first three periods of the periodic table having atomic weights below about 40. With this criterion and based on cost, availability and ease of handling, magnesium, aluminum, and silicon are materials which look attractive as mass quantity fuels for a nuclear-chemical energy cycle. Some discussion of the fuel processing cycles and possible types of converters for these fuels is also included.

I. INTRODUCTION

The utilization of the energy of nuclear fission requires the use of a relatively large and cumbersome apparatus involving a certain minimum quantity of fuel, regardless of the power, and a large heavy shield to contain nuclear radiations from the fission process. This is in contrast to chemical fueled energy sources, wherein the size of the energy releasing device (fuel burner) is proportional to the energy release rate throughout the whole size range. Thus, while a nuclear reactor may be comparable in bulk and weight (or even enjoy an advantage) to a chemical fueled thermal energy source in large size power supplies, the shielded nuclear supply becomes intolerably bulky for most small power unit applications. Hence, the direct application of nuclear heat to the whole of our energy

needs has been limited to those areas requiring the production of large blocks of power in very large size power units. The most readily exploited area in this connection is central station electric power generation. Since the size limitations on nuclear reactors are inherent and not subject to appreciable improvement, it is very likely that the release of nuclear fission energy will always be performed in large units.

Some perspective on our energy consuming habits is gained by examination of Table 1 which gives a tabulation of the energy-consuming sectors in the United States economy.

Table 1

THE RELATIVE IMPORTANCE OF ENERGY-CONSUMING
SECTORS IN THE U.S. ECONOMY, 1955⁽¹⁾

Sector	Percentage of Total Mineral Fuels and Hydropower Consumed
Industry	39.2
Commercial	8.5
Households	18.6
Transportation	20.2
Self-performed by households	9.3
Self-performed by gov't. (excl. military)	0.3
General	10.6
Government	4.6
Agriculture	1.8
Miscellaneous uses and losses	7.1
Total	100.0%
Consumed in the form of electricity	19.2%

From the data of Table 1 then, it is evident that nuclear power in the generation of electricity can supply a maximum of about one-fifth of our total energy requirements with the present pattern of energy consumption. Actually, the amount of nuclear power input for electricity would be somewhat less than this since the total electric power generated represents some 20% or so produced by hydropower and an additional quantity generated in quite small units which are too small for the application of nuclear reactors.

Current projections and estimates indicate that the world's supply of fossil fuels will be severely depleted and perhaps exhausted sometime within the next one-hundred years. Increased demands for energy coupled with a higher average standard of living and increases in population will result in a continuing rapid rise in the world's energy consumption.

Accompanying the increased demands for energy is a continued decrease in the supply of fossil fuels. The United States in the 1950's was consuming more than one-third of the world's energy supply.⁽²⁾ Reduced supplies of plentiful fuels are already being felt in the U.S. as evidenced by the fact that we changed from a net exporter of fuel to a net importer in the mid-1950's.

It seems inevitable, therefore, that a large portion of our energy needs will have to be supplied from nuclear sources in the future. As shown in Table 1, energy consumption for transportation is a large segment, amounting to about 20% of the total system input. The personal automobile (the listing - "Self-performed by households") accounts for almost one-half of total transport uses and 9.3% of the total energy consumed. In addition to being a sizable segment of total energy uses, transportation is the area most difficult for nuclear power application. All other segments of the economy, listed in Table 1, could, in principle, be supplied with energy in the form of electricity if it became really necessary and, thus, could be supplied with central station nuclear-electric power. (An exception is "Miscellaneous uses and losses," accounting for 7.1%, in which the energy generating capacity of the material is not the reason for its use. Rather, it is the physical and chemical characteristics other than combustibility that are exploited.) Transportation, on the other hand, implies portable power generating devices and, therefore, requires compact energy converters with a transportable supply of fuel. If the transportation segment of the economy is to be supplied by nuclear energy, a means for distributing the nuclear energy in a more easily handled form must be devised.

A method for distributing nuclear energy which appears to be attractive is storage by means of synthesizing chemical fuels in large processing plants using nuclear heat. Such a system would not differ radically from our present day transportation supply methods in that the fuels could then be dispensed to various points of distribution and made readily available in as small or as large a quantity as desired. One difference which might exist would be that the reaction products from the energy producing reaction in the converter would probably be retained and returned to the processing plant for regeneration. This is in contrast to most chemical fuels used today, wherein the reaction products are discarded to the atmosphere.

The following discussion, then, treats some of the factors involved in selecting chemical fuels for a nuclear-chemical energy distribution system. Section II of the report gives background information on the various energy conversion devices which are available for use in power supplies. Section III gives a brief resume of the parameters for radioisotope, nuclear, and selected chemical prime energy sources. Various energy source-converter combinations are discussed in Section IV. Finally, possible chemical fuels for a nuclear-chemical energy distribution system are explored in Section V and conclusions are given in Section VI.

II. COMPARISON OF VARIOUS ENERGY CONVERTERS

In considering the application of nuclear energy to small portable power supplies, it is necessary to review briefly the parameters of various energy converters which have been used in small power producing devices. By far the most widely used converter for transportation power plants is the internal combustion engine, of which the gasoline engine is the pre-dominant representative. For this reason, the performance of the gasoline internal combustion engine has been chosen as the standard for comparison of other converter-energy source combinations in Section IV of this report. Table 2 lists parameters for the various energy converters. The following discussion gives a brief description of the salient features of various converters. While it is recognized that some of the devices do not have application in automotive power units due to certain size restrictions, they are, nevertheless, included for comparison.

Table 2
SUMMARY OF PARAMETERS FOR VARIOUS ENERGY CONVERSION DEVICES

Conversion Device	Current Size Range	Specific Weight (kg/kw)	Power Density (kw/L)	Efficiency (%)	Operating Limits
1. Thermoelectric-lead telluride	several watts to 5 kw	60-20 ⁽¹⁾	0.015	5-15 ⁽¹⁾	Operating temperatures up to 540°C with present materials.
2. Thermionic converter	several hundred watts	15-1.0 ⁽¹⁾	0.10	10-30 ⁽¹⁾	Difficult mechanical problems with high temperature seals and close electrode spacings.
3. Fuel cells					
a. Hydrogen-Oxygen	up to 10 kw	125 ⁽³⁾ -15 ⁽²⁾	0.01-0.3	up to 80	Current densities limited by electrode polarization, particularly with gas electrodes.
b. Regenerative - LiH	several hundred watts	85	0.05	35 ⁽¹⁾	
c. Allis Chalmers petroleum gas ⁽⁶⁾	15 kw	75	0.02	60	
4. Photovoltaic cells	few milliwatts to 100 watts	10 ⁽⁴⁾ -1000 ⁽⁵⁾	0.2-0.002	10-20 ⁽¹⁾	Unfavorable surface-to-volume ratio, i.e., large surface area per unit power is required.
5. Heat engines					
a. Automobile engine	100 kw	4	0.50	20	Limited to large size units. >1.0 Mw in order to produce net power output - needs temperature in 3000-5000°F range.
b. Mercury turbine (For space power plant)	1000 kw	2	-	12	
c. Air-cooled aircraft	up to 5000 kw	1	-	25	
6. Magnetohydrodynamic (MHD)	11 kw (experimental device, not self-sustaining)	-	-	50% ⁽¹⁾ as a topping unit for steam cycle	

(1) Projected future performance

(2) High pressure cell

(3) Low pressure cell

(4) Unsupported solar cells

(5) Solar cells and supporting structure in an unoriented satellite - includes redundant cells to assure a continuous supply of power as satellite tumbles.

(6) A mixture of hydrogen and petroleum gases

A. Heat Engines

A typical passenger automobile engine of the 100-horsepower size, has a specific weight of around 4 kg/kw. The over-all efficiency of the engine is about 20%. In comparing the specific weight of this converter with those giving an electrical output, the use of the device must be considered. If the ultimate end use of energy is in the form of motion and mechanical force, as in the case of an automobile, the devices which produce an electrical output have a penalty due to the addition of the weight of electric motors. If the ultimate end use of the energy is as electrical power, then the automobile engine incurs a similar penalty due to the addition of the weight of an electric generator.

A lightweight mercury-cycle turbine producing a 1-Mw electrical output as described in Reference (3) has a specific weight, including the mercury inventory and electric generator, of 2 kg/kw. This turbine-generator has a rather low net cycle efficiency of 12% due to the fact that it is used in a power cycle designed to maximize radiator temperature and minimize radiator size in a space power plant. A central station steam turbine with an over-all cycle efficiency of perhaps 35% and no particular attempt being made to minimize weight would weigh about 18 kg/kw.

The highest power-to-weight ratio for an energy conversion device other than rocket or jet engines is given by an air-cooled aircraft engine. Such engines have specific weight of about 1 kg/kw and an over-all efficiency of about 25%.

B. Thermoelectric Devices

Although thermocouples have been used for years, thermoelectric power producing devices are in a very early stage of development. At present they are feasible for power supplies in the several hundred watt size range, and the largest size thermoelectric unit constructed so far is a 5000-watt power supply built for the U.S. Navy. Lead telluride is considered to be the only truly operational thermoelectric material available today. Other promising thermoelectrics that are currently being developed are cadmium sulphide, cobalt silicide, and gadolinium selenide. These materials offer Seebeck coefficients comparable to that of lead telluride, but high temperature performance as well.^(4,5)

Thermoelectric materials have been used in the SNAP series of generators for space applications and will be used in future units of this type now under development having power outputs up to 125 watts. Thermoelectric converters are also being used in power supplies for remote arctic weather relay stations and also in navigational buoys currently under development. The commercial applications of thermoelectric devices have been limited, although recently several thermoelectric cooling devices including

an office water-cooler and a small refrigerator have been put on the market. Another commercial application utilizes thermoelectric elements for powering an air circulation fan in a gas-fired home heating unit.

The efficiency of thermoelectric devices runs about 5% today, but they are believed to be capable of ultimately yielding efficiencies of about 15%. These higher efficiencies will be obtained through the development of thermocouples capable of operating at higher temperatures. Recent life testing of lead telluride devices have been somewhat discouraging in this respect, and results indicate that the upper temperature limit should be reduced from 1100°F to 800°F or 900°F for long-life operation.^{(4)*} The specific weight of current thermoelectric devices is about 60 kg/kw and may be reduced to perhaps 20 kg/kw with improved performance materials.

C. Thermionic Devices

Thermionic conversion devices are somewhat behind the thermoelectrics since the converters built to date have all been proof of principle or experimental devices and have not been constructed for any specific useful power application. The maximum power output on these experimental devices has been about 100 watts.

An ideal thermoelement should have a good electrical conductivity and a low thermal conductivity. Thermionic devices approach this ideal more closely than do the thermoelectric devices and, as a result, may be expected to yield higher efficiencies. The efficiencies of experimental devices built so far have been as high as about 15%. It is expected that future practical thermionic converters will give efficiencies up to 30%.⁽⁶⁾ A current thermionic converter would have a specific weight of about 15 kg/kw and, with expected improvements, this might be reduced to 1.0 kg/kw in future devices.

Thermionic devices also have the advantage that it is feasible to convert the electrical output from d-c to a-c directly in the device by the use of a small grid voltage. In considering the application of thermionic converters to nuclear power sources, there may be some beneficial effect from the nuclear radiations in that they can be used to reduce the space charge.

D. Magnetohydrodynamic (MHD) Devices

The magnetohydrodynamic converter system is in an early stage of development. Feasibility has been demonstrated with an 11-kw generator built by the Avco-Everett Research Laboratory.⁽⁷⁾ In order to obtain sufficiently high gas electrical conductivity, very high gas temperatures are

* Advances are being made on alternate materials. The Radio Corporation of America reports in the December 7, 1961, issue of Machine Design that they have developed a thermoelectric material which will operate at 1800°F, thereby giving higher efficiencies.

required in the generator. However, due to the absence of hot, highly stressed, moving parts, a MHD generator should be able to handle high temperatures thereby permitting high thermodynamic efficiencies. Calculations by workers at Avco indicate that MHD generators may be considerably lighter in weight than conventional turbine generators. MHD devices require temperatures greater than 3000°F and, obviously, the attainment of such high temperatures in continuously operating units requires significant advances in materials technology.

If MHD systems are to be used as central station converters, they would probably be used as topping devices, with the reject heat from the MHD portion of the plant going to a conventional turbine-generator. A high heat reject temperature of the MHD generator is a requirement since the device depends on thermal ionization of the gas to maintain a high gas electrical conductivity. The MHD generator is limited to sizes greater than about one megawatt. This results from the fact that in small size generators the magnetic field excitation requirements and the heat transfer losses exceed the electrical output of the generator. Thus it does not appear that MHD generators will have application as small power supplies.

E. Fuel Cells

Although the fuel cell is over one-hundred years old, attempts to build a practical and workable cell were unsuccessful until a few years ago. Basically the fuel cell is a continuous feed primary battery and the materials which react in the cell to produce an EMF can be continuously replaced as the reaction proceeds. This means that the operating lifetime of the cell is no longer limited by the amounts of reacting materials which can be stored in the cell proper as in the case of conventional batteries. When considering the fuel cell as an energy conversion device, the most frequently mentioned characteristics are the high conversion efficiency and lack of moving parts. Since a fuel cell operates isothermally and escapes the Carnot limitation of a cycle involving temperature changes, efficiencies greater than 90% are theoretically possible for some types of cells. Of course, actual efficiencies will always be less than theoretical since the cells are subject to losses due to electrode polarization and internal resistance as are conventional batteries. Also, particularly in high-temperature cells, losses in efficiency result from undesirable side reactions and heat losses. Even so, practical efficiencies as high as 80% have been obtained with some types of cells.

The current development of fuel cells is directed toward practical operating systems from several watts up to about 100 kw. Most of this development in the United States is government financed with the objective of military application. The most highly developed fuel cell system at the present time is the hydrogen-oxygen cell, wherein hydrogen and oxygen gas are fed to respective electrodes immersed in an electrolyte solution. The Union Carbide hydrogen-oxygen cell⁽⁸⁾ operates at 70°C and atmospheric pressure and is an example of the low-pressure type of cell. The Bacon

high-pressure cell⁽⁹⁾ operates at 200°C and 40 atm. Representative specific weights for hydrogen-oxygen cells are 125 kg/kw for the low-pressure, low-temperature cell and 15 kg/kw for the high-pressure cell. To date, the hydrogen-oxygen cells have been constructed in multicell units ranging up to about 10 kw.

Another class of fuel cells is represented by the Allis-Chalmers petroleum gas cell.⁽¹⁰⁾ This cell operates at low pressure and a relatively low temperature. The fuel is a mixture of petroleum gases and the oxidant is oxygen.* As in the case of the hydrogen-oxygen cell, the electrode reactions are accelerated by catalysts on the electrode surfaces. This type of cell has been built in a large 15-kw multicell unit and used in a demonstration to power a tractor driven by electric motors. The specific weight of the cell is 75 kg/kw and the efficiency is reported as 60%.

Another type of fuel cell which deserves mention is the thermally regenerative cell. The most promising to date in this category is the lithium hydride cell proposed originally by Mine Safety Appliance Research Corporation.⁽¹¹⁾ In the MSA cell, liquid lithium and gaseous hydrogen are reacted to form lithium hydride. Lithium hydride is unstable at elevated temperatures and decomposes to give hydrogen and lithium. By removing the lithium hydride reaction product from the cell electrolyte and heating it to an elevated temperature, it is possible to regenerate the individual reactants - lithium and hydrogen. Development work on this fuel cell has also been carried out by the Thompson Ramo Wooldridge Company and they have built a multicell unit with an output of 500 watts. The specific weight of the TRW cell is 85 kg/kw. So far the lithium hydride fuel cell has been operated in the Laboratory by feeding lithium and hydrogen continuously to the cell. The feasibility of a continuous cyclic regeneration process to reform hydrogen and lithium is yet to be demonstrated. The electrolyte for the lithium hydride cell is a mixture of fused salts. The operating temperature is above the melting point of the salts and is upwards of 400°C.

Perhaps the best performance in terms of weight per unit of electrical output and power density is obtained with the fuel cell systems using metallic fuels. Representative of this type of fuel cell are the zinc-chlorine cell and the sodium-oxygen cell. In the sodium-oxygen cell,^(12,13) sodium and oxygen are reacted at their respective electrodes to form sodium hydroxide. The electrolyte is a solution of sodium hydroxide. Because of the great reactivity of sodium with water, it is necessary to feed the sodium as an amalgam containing about 0.5 wt-% sodium. This cell is under development for the Navy's Bureau of Ships with the objective of producing a prototype power plant which will develop 75 kw. The specific weight of the cell has been reported as 7 kg/kw. This weight-to-power ratio is the best that has currently been reported for a fuel cell.

* The operational tests to date with this cell operating on mixtures of petroleum gas and hydrogen have been discouraging. However, the cell gives satisfactory performance when operated on pure hydrogen.

F. Photovoltaic Devices

When the silicon cell was first announced in 1954 by its inventors at the Bell Telephone Laboratories, the nominal solar energy conversion efficiency (at room temperature) was given as 6%. This was about ten times better than the selenium cell, the best previously known photovoltaic converter. Since then, continued research and development has raised the conversion figure of merit to 14%, with production quantities of cells available in the range of 10-12%. Because of the nature of light and of silicon, the theoretical maximum efficiency is estimated to be less than 20%. The spectral response of silicon solar cells lies mainly in the visible and near infrared regions of the solar spectrum, peaking at about 0.8 microns. Incident radiation of wave length shorter than about 1.1 microns is capable of being absorbed in the top portion of the solar cell and converted directly into electricity.⁽¹⁴⁾ Solar cell power units have been built in sizes ranging from several milliwatts up to about 100 watts. The six-inch Vanguard satellite used a solar cell power supply which supplied about 40 milliwatts.

The specific weight of an unsupported cell in direct sunlight is 10 kg/kw for a 12% efficient cell. This is an ideal figure and can be considered to be the best that is obtainable with cells currently available. Indeed, these figures are reduced considerably in some applications. For example, the specific weight of a solar power supply in an unoriented satellite in sunlight 100% of the time with no storage would be 1000 kg/kw. The weight increase is due to reduction in efficiency by protective plates for the cells, structural materials to support the cells, and a multiplicity of supplies to assure a continuous power output as the satellite tumbles. Of course, some of these provisions would not be required in an earth-bound power supply and a specific weight of 50 to 100 kg/kw for such a terrestrial solar power supply might be expected.

An inherent feature of solar power supplies is their required high surface-to-volume ratio. A 100-watt supply containing 10,600 silicon cells constructed by the International Rectifier Corporation consists of a panel measuring about 1.5 meters on a side. This large surface area requirement is a disadvantage for small size mobile power supplies. It may even be a limiting factor in large power generation units located in desert areas where in the losses due to collection of power over a large area may become inordinately high. Such factors must be considered when examining specific applications for solar cells.

G. Energy Storage Devices

The term "energy storage" implies the storage of energy by means of some device so that it may be released readily in a desired form at some future time. By this definition transportation power supplies are also energy storage devices, since the energy converter and fuel supply constitute a unit

which can give power at some future time when needed. The energy storage capabilities of some devices which do not involve heat conversion but store energy, either in its ultimately used form or as chemical energy, are tabulated in Table 3.

Table 3

PARAMETERS FOR ENERGY STORAGE DEVICES*

<u>Batteries</u>	<u>Specific Energy (kcal/g)</u>
Lead acid	0.030
Ruben mallory (mercury cell)	0.091
Zinc-silver oxide	0.122
Edison cell (nickel-iron)	0.028
Lelanche (dry cell)	0.047
<u>Mechanical</u>	
Springs	6.0×10^{-5}
Flywheels	4.7×10^{-3}

* Source: Reference 15

Note: to convert values in Table to watt-hr/kg
multiply by 1163.

When speaking of such devices, an important figure of performance is the energy storing capability in energy units per unit weight. There are many ways of storing energy such as charging capacitors, pumping water uphill and winding springs, but the most convenient and efficient method for a wide variety of uses is the electric battery. Unfortunately, batteries although they are better than many alternate devices, have relatively limited energy storage capabilities. Furthermore, they are expensive and the storage of large quantities of energy by this method is very costly.

One of the factors limiting the storage capacity of batteries is the rather low material efficiencies which can be attained, i.e., the quantity of reactant present in the battery electrodes is several times the amount which actually reacts before the battery is completely discharged. The fuel cell circumvents this problem by continuously feeding reactants to the cell and thereby gives material efficiencies of close to 100%.

III. COMPARISON OF VARIOUS HEAT SOURCES

The gasoline engine and the fuel cell are converters which require no external apparatus for burning the fuel, i.e., the fuel reacts within the device and the output is mechanical energy in the form of torque in one case or electrical energy in the other. Therefore, either of these devices requires energy input in the form of a chemical fuel suitable for the specific requirements of each converter. Thermoelectric, thermionic and indirect cycle turbine power plants, on the other hand, depend upon energy input in the form of heat, and thus require additional apparatus to supply this heat. Therefore, it is possible to consider the application of nuclear or radioactive isotope heat in addition to chemical heat for these converters. In order to compare the chemical fueled systems with isotope or nuclear reactor powered systems, it is necessary to know parameters for these various fuel supplies.

A. Isotope Heat

The weight-to-power ratios for some radioactive isotopes which have been considered for heat sources ranges from about 8.9 kg/kw for strontium-90 to a value of 0.0075 kg/kw for polonium-210. Table 4 summarizes the power output and half-life data for a number of radioisotopes.

Table 4

TYPICAL RADIOISOTOPE HEAT SOURCES*

Nuclide	Mode of Decay	Half Life	Fuel Form	Density (g/cm ³)	Thermal Power (watt/cm ³)	Specific Weight (kg/kw)	Estimated Costs (\$/watt)
Po ²¹⁰	Alpha	138 d	Po	9.3	1320	0.0075	-
Cm ²⁴²	Alpha	162 d	Cm ₂ O ₃	11.8	1169	0.010	45
Pu ²³⁸	Alpha	86.4 yr	PuC	12.5	6.9	1.8	1600
Ce ¹⁴⁴	Beta	285 d	CeO ₂	6.4	12.5	0.51	14
Pm ¹⁴⁷	Beta	2.6 yr	Pm ₂ O ₃	6.6	1.1	6.0	1630
Cs ¹³⁷	Beta	33 yr	CsCl	3.9	1.27	3.1	54
Sr ⁹⁰	Beta	28 yr	SrTiO ₃	4.8	0.54	8.9	23

* Source: Reference 5

From the projected costs listed in the last column of Table 4, it is seen that radioisotopes are indeed very expensive heat sources. Furthermore, it appears that they will always be expensive since the artificially produced isotopes require irradiation in reactors, and the isotopes obtained from fission products require expensive chemical processing to separate

them. It has been stated that a value 0.10 watts/g is the minimum useful specific power for a pure radioisotope.⁽¹⁶⁾ Thus far, the use of mixtures of fission products has resulted in a maximum of only 0.01 watts/g due to the decay of short-life fission products. Therefore, prospects for a cheap gross fission product heat source are discouraging.

Another disadvantage of isotope heat sources is the fact that the power supply operates at full capacity at all times regardless of demand loads. This is undesirable for applications where the power supply would be operated intermittently. Also, the total supply of radioisotopes from fission products is dependent upon the installed capacity of nuclear power plants and, hence, is probably limited to relatively small quantities for some time to come.

B. Nuclear Reactor Heat

In considering the use of nuclear reactors as heat sources for small mobile power supplies, the reduction of shielding weight to a reasonable value presents one of the major problems. In the mobile nuclear reactor power plants considered thus far where weight is a limiting factor, such as the Aircraft Nuclear Reactor, the SNAP series of space power reactors, and the Lunar Power Reactor,⁽³⁾ the approach has been to provide very limited shadow-type shielding or, where possible, to eliminate shielding entirely. An example of this latter approach is the LPR wherein the reactor would be set into a hole in the moon's surface. Figure 1 shows the specific weight (kg/kw) plotted versus reactor output (kw) for the SNAP series of reactors and the LPR. The data of the curve includes no weight for shielding. Included on the figure for comparison are the specific weights of some of the radioisotope heat sources.

? — The SNAP reactors⁽¹⁷⁾ are zirconium hydride moderated, using a homogeneous mixture of zirconium hydride and uranium for fuel elements. Consequently, below about 1000 kw the reactors are criticality limited and the specific weight shows a sharp reduction with reactor output up to this power level. Above this power level, the reactors are heat transfer limited and the reduction in specific weight with reactor output becomes much more gradual. Since they are criticality-limited, it is not possible to reduce the weights of these small reactors greatly. Some reduction might be possible should it be advisable and reasonable to use small fast reactors. In any event, the fast reactor systems offer gains in the larger size power plants since their power densities are higher. The penalty imposed by shielding in the small size reactors may be illustrated by the fact that the SNAP-2 (50 kw) reactor weight would be increased by a factor of 100 or so if continuous personnel access were required.

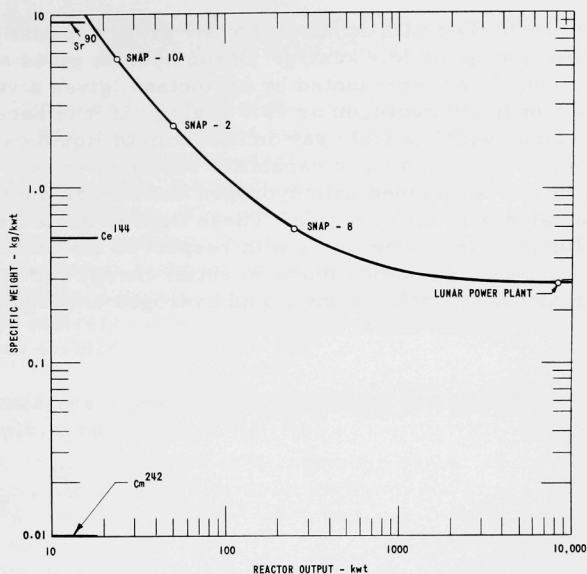


FIG. 1
SPECIFIC WEIGHTS OF REACTOR HEAT SOURCES AND ISOTOPE HEATERS
(NO SHIELDING)

C. Chemical Fuels

When comparing chemical power with nuclear reactor or radio-isotope heat sources, an important parameter is the energy available per unit weight of chemical fuel. Table 5 is a tabulation of the specific energy storage for a number of chemical fuel systems.

Table 5

SPECIFIC ENERGY OF SOME CHEMICAL FUEL SYSTEMS

Fuel	Oxidant	Specific Energy of Fuel System* (kcal/g)
Liquid hydrogen	Atmospheric oxygen	28.9
Boron hydride (B_5H_9)	Atmospheric oxygen	17.0
Iso-octane	Atmospheric oxygen	10.6
Liquid hydrogen	Liquid oxygen	3.21
Iso-octane	Liquid oxygen	2.36
Iso-octane	Oxygen gas in pressure tanks	0.294
Hydrogen gas in pressure tanks	Oxygen gas in pressure tanks	0.120

* Includes weight of fuel, oxidizer (except where atmospheric oxygen is used), and tankage (where materials are carried as gases).

Liquid hydrogen burned with atmospheric air gives the best performance with a specific energy of 28.9 kcal/g. Boron hydride gives a value of 17.0 kcal/g. Gasoline, represented by iso-octane, gives a value of about one-third that of liquid hydrogen or 10.6 kcal/g. If it is necessary to carry the oxidizer along with the fuel, say in the form of liquid oxygen, corresponding reductions in energy storage capability result as shown. The poorest storage capability is obtained with hydrogen and oxygen as gases in pressure tanks with a value only 0.120 kcal/g. These figures show the very favorable position of liquid hydrocarbon fuels with respect to the others. A passenger automobile carries three times more potential energy per unit weight of fuel than does an advanced rocket using liquid hydrogen and liquid oxygen fuels.

IV. COMPARISON OF SOME HEAT SOURCE- CONVERTER COMBINATIONS

The parameters for some of the conversion devices and fuels are combined to roughly illustrate what systems have the best performance in terms of minimum weight requirements for different duration missions. For purposes of reference, the application chosen is that of the conventional gasoline automobile engine. The power output of such a unit is of the order of 100 hp or 75 kw. In making the comparisons, it is necessary that the final output of the power plant be mechanical force; therefore, the electrical systems (solar cells, batteries, etc.) require an electric motor. The nuclear system uses an indirect cycle liquid-metal-cooled reactor with a heat exchanger and gas turbine.

If we consider a power plant consisting of a converter or heat source and a fuel supply, then,

$$\frac{1}{E} = \frac{1}{\eta E_f} + \frac{W_c}{\ell} \quad (1)$$

where,

E = over-all specific energy of converter and fuel, kcal/g,

η = thermal efficiency of converter,

ℓ = duration of mission, hr,

W_c = summation of specific weight terms for converter and heat source, g/(kcal/hr) of output,*

E_f = specific energy of fuel, kcal/g

Figure 2 contains plots of Eq. (1) for a series of systems. The values for the specific weights of converters and heat sources are taken from Table 2 and Fig. 1. The specific energies of chemical fuels are taken from Table 5. The over-all efficiency for the automobile engine is 20%. The hydrogen-oxygen fuel cell was assumed to have an efficiency of 70%. The specific weight of the cell was taken as 15 kg/kw, representative of the best which might be attained with high pressure cells. A specific weight of 100 kg/kw was used for the solar cells, intermediate between values for unsupported cells and actual values obtained in the Vanguard satellite.

The specific weight of the unshielded reactor heat source plus heat exchangers and gas turbine is 8 kg/kw. The over-all efficiency of the cycle is 25%.

*g/(kcal/hr) = 1.162 x kg/kw.

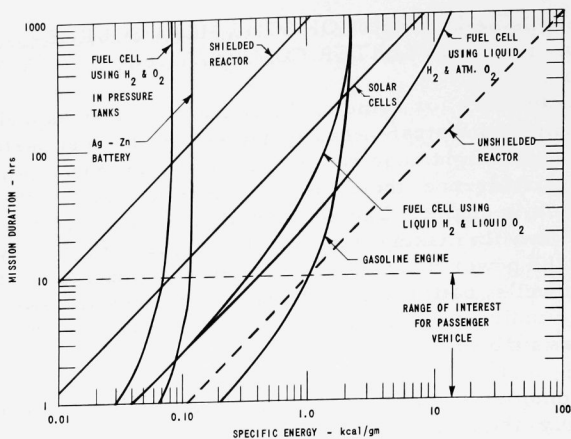


FIG. 2
SPECIFIC ENERGY vs MISSION DURATION FOR
SEVERAL CONVERTER - FUEL SYSTEMS

Since the use of an unshielded reactor is impossible for application as a passenger vehicle power supply, the curve is included only for comparison. The specific weight of printed circuit electric motors for use with the battery, fuel cell, and solar cell power supplies is 6 kg/kw.

As seen from the form of Eq. (1) and the curves of Fig. 2, the weight of the converter and/or heat source becomes less important for the chemical-fueled systems as the duration of mission increases and the specific energy of the power plant plus fuel supply asymptotically approaches the product of the converter efficiency and the specific energy of the fuel. Thus, the high pressure hydrogen-oxygen fuel cell using liquid hydrogen and oxygen is about equivalent to the gasoline engine for durations greater than 100 hr, since the products of their respective efficiencies and fuel specific energies both equal about 2 kcal/g.*

The devices for which the duration of operation is limited by the lifetime of the converter, i.e., nuclear reactors and solar cells show a linear increase in specific energy with mission duration. The longer they can be operated, the higher will be the specific energy of the system, and limits are set by this lifetime rather than the properties of a fuel.

*The assumption made here is that cryogenic storage of hydrogen and oxygen requires the same percentage weight as tankage for gasoline. While it is recognized that this is by no means the case, these comparisons were made mainly from the viewpoint of the inherent limitations of the various chemical fuels and thus define upper limits.

Of course, there is an upper limit to the amount of energy stored in a given mass of nuclear fuel, but considerations of reactivity loss, etc., will dominate for a nuclear reactor heat source.

For application to passenger vehicles, the mission durations up to 10 hr or so are of interest. The curves of Fig. 2 are best used as an indication of the relative positions of the various power plants. The high pressure hydrogen oxygen fuel cell is within a factor of two of gasoline engine performance. Needless to say, considerations of practicality have been set aside in making this comparison since the transport of liquid hydrogen and oxygen as fuel in a passenger vehicle is not currently feasible.

Neither is the unshielded reactor practical for such an application and, again, it is included for comparison. It is estimated that addition of adequate shielding will increase reactor weight by a factor of 100 or so. This, then, reduces the nuclear reactor to the poorest position of all the systems considered.

If the solar cells provide the steady state automotive power supply output of 75 kw alone (without storage), the area required is about 1680 sq meters or an area 41 meters on a side. This is unreasonably large and even if the projected maximum efficiencies of the cells are attained, an improvement of only a factor of 2 or so results.

The hydrogen-oxygen fuel cell combined with fuel supplies as gases in steel pressure tanks gives performance approaching the best chemical batteries, but still a factor of 5 below that obtainable if the hydrogen and oxygen are liquified. This weight penalty for tankage applies to gaseous electrode fuel cells in general, and methods such as liquification or other compact storage must be adopted if their performance is to greatly exceed that of the best conventional batteries.

The foregoing discussion has shown the relative performance of a series of fuel-converter combinations in terms of the weight required to store a given quantity of energy for missions of various durations. It is evident that the direct application of nuclear power to small portable power supplies for surface transportation is not feasible. Storage of energy in chemical fuels is a necessary feature of future distribution of nuclear power for small automotive power supplies. The remaining sections of the report are devoted to a discussion of the possibilities and limitations of various chemicals which might be successful alternates to hydrocarbon fuels.

V. FUELS FOR USE IN A NUCLEAR-CHEMICAL ENERGY DISTRIBUTION CYCLE

A. Characteristics of a Desirable Chemical Fuel for Automotive Transportation

In order for a chemical to be used widely in surface transportation power plants and handled on a routine basis, there are certain criteria as to energy storage capability, toxicity, stability, and abundance of fuel supply source which must be met. While many of these requirements are obvious, they are nevertheless reviewed here as an aid to the later discussions.

1. Energy Storage Capability

In order for a fuel to be considered at all it must have a reasonable minimum energy storage capability. For purposes of comparison, the most common fuel for transportation use today namely, gasoline, has a storage capability of 10.6 kcal/g or 7.5 kcal/cm³. These are values which should be kept in mind in considering alternate chemical fuels. The relative importance of the energy per unit weight and energy per unit volume figures depend on the particular application involved. For surface transportation missions, the storage per unit volume is probably the more critical parameter since, in general, a two or three-fold increase in the weight of fuel could be tolerated, whereas corresponding increases in the volume required could cause problems. Conversely, in applications such as aircraft the total weight of the fuel is probably the more important consideration.

2. Chemical Characteristics of Fuel

In order for a fuel to be widely used on a routine basis, it must be relatively easy and convenient to store for long periods of time and be readily dispensed when needed. Furthermore, it should be nontoxic and reasonably unreactive with structural materials and the atmosphere at ordinary temperatures. A further desirable characteristic is that the fuel be readily oxidized by atmospheric oxygen and/or be capable of undergoing an electrochemical reaction in a fuel cell.

3. Chemical Characteristics of Fuel Reaction Products

The same general requirements as to toxicity and chemical stability for the fuel apply to the reaction products of the energy producing reaction. In the case of the hydrocarbon fuels, the reaction products, of course, are volatile and are exhausted to the atmosphere. Disposal of combustion products in this manner is not necessarily a requirement for chemical fuels in the nuclear-chemical fuel system and, in fact, this may

be undesirable from a material efficiency standpoint. A corollary is, if the reaction products are to be retained, they must be in the form of liquids or solids, rather than gases. If they are gases, they must be of such nature that they can be exhausted into the atmosphere without danger.

4. Natural Abundance of Fuel Supply Source

The fuel raw material must be available in sufficient quantity and concentration to provide the large quantities of fuel required to support our transportation system. The total quantities in the earth's crust of most materials that one might consider are sufficient but, rather, it is the lack of concentrated deposits which is usually limiting. In this connection, the regenerative fuel cycle wherein the converter reaction products are retained and recycled to the nuclear processing plant is attractive because of a good material efficiency. However, in the case of a raw material which is readily available and accessible, e.g., water, a high material efficiency is not particularly important.

B. Survey of Elements and Compounds for Use as Fuels

In selecting suitable materials for use as fuels, consideration has first been given to the energy storage capability of the material, since listing of this parameter for a number of materials very quickly narrows down the available selections. In the chemical binding of atoms, the amount of energy released or absorbed is proportional to the number of chemical bonds formed or severed, for a given type of bond. Consequently, as might be expected, the formation of compounds between the lighter atoms give greater amounts of energy per unit weight of material than those formed between larger or heavier atoms. Therefore, the materials of interest for high energy storage fuels are the elements in the first few periods of the periodic table.

The good storage capability of hydrocarbon fuels is due to the fact that they are composed of two of the lightest elements, hydrogen being first, and carbon number six in the periodic table. It is evident that an alternate fuel cannot stray too far from these positions in the periodic table if it is to give performance approaching that of a hydrocarbon fuel. A further advantage of hydrocarbon fuels is that they can be oxidized readily by the oxygen in air, thus eliminating the need for carrying with the converter one constituent of the energy producing chemical reaction. This results in a considerable saving in fuel weight as evidenced by the fact that a molecule of carbon dioxide contains only twenty-seven per cent carbon by weight and a molecule of water contains only eleven per cent of hydrogen by weight. It is desirable for an alternate chemical fuel to take advantage of the atmospheric oxygen in the same manner, if possible.

Figure 3 shows the energy storage capability plotted versus the atomic weight of the element for a series of light element oxides and

nitrides. As can be seen from the figure, there is a sharp drop-off in energy storage capability with increasing atomic weight. Also, the energy storage increases with increasing quantity of oxygen or nitrogen in the oxide or nitride combustion product (increasing number of bonds formed). The heats of formation of nitrides are considerably lower than the corresponding oxides. Hence, the formation of nitrides as reaction products should be avoided for maximum fuel performance.

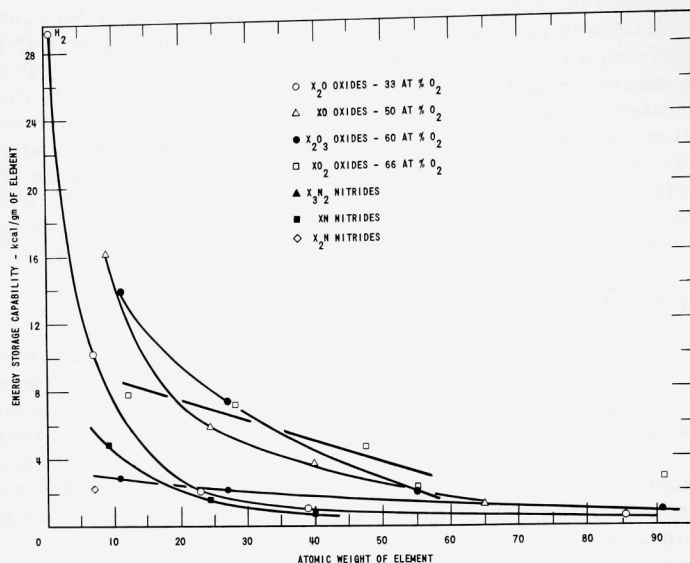


FIG. 3
ENERGY AVAILABLE BY OXIDATION AND NITRIDING OF LIGHT ELEMENTS

Table 6 gives energy storage data for the light elements and light element hydrides up to and including calcium (atomic weight - 40.1), for the formation of their oxides. The columns labeled W_c/W_f and V_c/V_f give the ratio of the weights and volumes of combustion products to the weight and volume of the original fuel. These ratios are of interest in considering the regenerative type of fuel cycle wherein the reaction products would be retained in separate tanks for return to the processing plant for regeneration. In such a cycle, the oxidizer may be atmospheric air, but if the combustion products are retained, the total weight of combustion products stored when all the fuel is spent is equal to the weight of fuel plus the oxygen used to burn it. In this case, the saving in weight due to the using of atmospheric oxygen is not too meaningful since the vehicle must have provision for storing this additional weight as fuel is oxidized. The convenience of not requiring storage space for oxidizer as a prime fuel remains, however.

Table 6

ENERGY STORAGE FOR LIGHT ELEMENTS AND COMPOUNDS BY FORMATION OF THEIR OXIDES

Chemical Fuel	ΔH per Unit Wt. of Fuel (kcal/g)	Fuel Density (g/cm ³)	ΔH per Unit Vol. of Fuel (kcal/cm ³)	Oxidation Products	ΔH per Unit per of Combustion Products (kcal/g)	$\frac{W_c^*}{W_f}$	$\frac{V_c^{**}}{V_f}$
H ₂	28.9	0.071(L)	2.05	H ₂ O	3.21	9.0	0.639
B ₅ H ₉	16.2	0.61	9.9	B ₂ O ₃ , H ₂ O	4.02	4.03	1.69
Be	16.1	1.85	29.8	BeO	5.81	2.77	1.70
B	13.9	2.32	32.4	B ₂ O ₃	4.33	3.21	4.04
C ₈ H ₁₈ (gasoline)	10.6	0.703	7.46	CO ₂ , H ₂ O	4.59	Volatile Products	
Li	10.3	0.53	5.30	Li ₂ O	4.79	2.15	0.567
LiH	9.74	0.78	7.59	Li ₂ O, H ₂ O	3.23	3.02	1.61
LiAlH ₄	9.55	2.55	23.4	Li ₂ O, Al ₂ O ₃	3.54	2.70	3.80
PH ₃	7.90	0.75(L)	5.89	P ₂ O ₅ , H ₂ O	2.74	2.88	1.27
C	7.83	2.0	15.7	CO ₂	2.13	Volatile Products	
Al	7.39	2.7	20.0	Al ₂ O ₃	3.93	1.88	1.28
Si	7.20	2.0	14.4	SiO ₂	3.36	2.14	1.84
Mg	5.91	1.74	10.3	MgO	3.56	1.66	0.806
P	5.81	1.82	10.6	P ₂ O ₅	2.54	2.29	1.74
NH ₃	3.99	0.817(L)	3.26	NO ₂ , H ₂ O	0.928	Volatile Products	
Ca	3.78	1.55	5.86	CaO	2.70	1.40	0.653
H ₂ S	3.63	~1(L)	3.63	SO ₂ , H ₂ O	1.51	Volatile Products	
N ₂ H ₄	3.49	1.01	3.52	NO ₂ , H ₂ O	0.871	Volatile Products	
NaH	2.69	1.40	3.77	Na ₂ O, H ₂ O	1.61	1.67	1.32
S	2.21	2.05	4.52	SO ₂	1.11	Volatile Products	
Na	2.16	0.97	2.09	Na ₂ O	1.60	1.35	0.576
KH	1.54	1.45	2.23	K ₂ O, H ₂ O	1.11	1.39	1.05
K	1.10	0.86	0.946	K ₂ O	0.916	1.20	0.445

$$^* \frac{W_c}{W_f} = \frac{\text{weight of combustion products}}{\text{weight of fuel}}$$

$$^{**} \frac{V_c}{V_f} = \frac{\text{volume of combustion products}}{\text{volume of fuel}}$$

As can be seen from the table, hydrogen, the lightest element, has the best storage capability per unit weight, but, due to the very low density of liquid hydrogen, requires greater volume than most other fuels. As would be expected, only the very lightest elements and their hydrides, namely lithium, beryllium, and boron (beryllium does not form a hydride) give greater energy storage than gasoline. Of the remaining elements and hydrides listed in the table, most have a performance on a weight or volume basis at least equal to half that of gasoline or better.

Table 7 lists the melting and boiling points of the various oxides, nitrides and fluorides of the light elements. As can be seen from this data, the only fuel materials other than hydrocarbons giving volatile oxide combustion products are hydrogen, carbon, hydrogen sulfide, sulfur, ammonia and hydrazine. A wide variety of possibilities for forming different fuels exists by combining various elements listed in Table 7. In this manner, it may be possible to form a fuel with more desirable physical properties for handling, etc. If the reaction between the two elements

is exothermic, however, then the resulting storage of energy in the compound on subsequent oxidation will be slightly lessened since the exothermic heat of combination will have to be supplied by the combustion reaction in decomposing the elements. If the combination is endothermic, the energy storage will be increased since this energy becomes available in addition to the heat of combustion on decomposition of the elements.

Table 7
PROPERTIES OF OXIDES, NITRIDES AND FLUORIDES OF LIGHT ELEMENTS*

Element	Atomic Weight	Oxides			Nitrides			Fluorides		
		Formula	Melting Point (°C)	Boiling Point (°C)	Formula	Melting Point (°C)	Boiling Point (°C)	Formula	Melting Point (°C)	Boiling Point (°C)
H ₂	1.008	H ₂ O	0	100				HF	-92.3	19.4
Li	6.94	Li ₂ O	>1700	-	Li ₃ N	<100	(d)	LiF	870	1676
Be	9.01	BeO	2530	~3900	Be ₃ N ₂	2200 ± 100	2240 (d)	BeF ₂	800	-
B	10.82	B ₂ O ₃	~450	1500	BN	3000 (s)	-	BF ₃	-127	-101
C	12.01	CO	-207	-190				CF ₄	-184	-128
		CO ₂	-56.6 (5.2 atm)	-78.5 (s)						
N	14.01	N ₂ O	-102.4	-88.49				NF ₃	-216.6	-120
		NO	-163.6	-151.8						
		N ₂ O ₃	-102	3.5 (d)						
		NO ₂	-9.3	21.3 (d)						
		N ₂ O ₅	30	47 (d)						
		NO ₃ (d slightly at ordinary temperature)								
F	19.00	F ₂ O	-	-167						
Na	22.99	Na ₂ O	-	1275 (s)	Na ₃ N	300 (d)		NaF	980-997	1700
		Na ₂ O ₂	460 (d)	657 (d)						
Mg	24.32	MgO	2800	-	Mg ₃ N ₂	1500 (d)		MgF ₂	1396	-
		MgO ₂	-	-						
Al	26.97	Al ₂ O ₃	2050	2250	AlN	>2200	2000 (s)	AlF ₃	1040	-
Si	28.06	SiO ₂	1710	2230	Si ₃ N ₄	1900 press; (s)	-	SiF ₄	-77	-65 (181 mm)
P	30.98	P ₂ O ₃	23.8	173	P ₃ N ₅	800 (d)		PF ₃	-160	-90
		P ₂ O ₄	>100	180 (s)						
		P ₂ O ₅	563	347 (s)						
S	32.07	SO	(d)	(d)	S ₄ N ₂	11	(d)	SF ₆	-50.8 (under pressure)	-63.5 (s)
		S ₂ O ₃	75-95 (d)	-	S ₄ N ₄	179 (s)	160 (e)			
		SO ₂	-75.5	-10.0						
		SO ₃	16.8	44.8						
		S ₂ O ₇	0	10 (s)						
		SO ₄	0-31 (d)	-						
Cl	35.46	Cl ₂ O	-20	3.8	ClN ₃	-	-	-	-	-
		ClO ₂	-59	9.9						
		ClO ₄	-	(d)						
K	39.10	K ₂ O	-	-	K ₃ H	(d)	-	KF	880	1500
		K ₂ O ₂	490	-						
		K ₂ O ₃	430	-						
		KO ₂	~400	(d)						
Ca	40.08	CaO	2580	2580	Ca ₃ N ₂	900	-	CaF ₂	1360	-
		CaO ₂	275 (d)	-						

*Source: Handbook of Chemistry & Physics

(d) - decomposes; (s) - sublimates; (e) - explodes

The possibility of using the exothermic heats of formation of some intermetallic compounds with the light elements has also been considered.

However, as shown by the data of Table 8, heats of formation of the intermetallics tend to be much lower than the formation of corresponding metal oxides, and hence, these reactions give a much lower energy release per weight of reactants than the oxidation reactions.

Table 8

HEATS OF FORMATION OF SOME INTERMETALLIC COMPOUNDS*

Compound	Heat of Formation ($-\Delta H$)		Compound	Heat of Formation ($-\Delta H$)	
	kcal/mol	kcal/g		kcal/mol	kcal/g
K ₂ Se	79.3	0.505	Al ₂ Fe	20.0	0.182
Na ₂ Se	63.0	0.504	Na ₃ Bi	45.6	0.166
SrSe	83.0	0.495	HgLi	21.0	0.101
Al ₃ Ca	56.0	0.463	Hg ₂ Li	25.0	0.061
AlNi	34.0	0.396	K ₂ Na	5.7	0.056
Li ₄ Sn	47.0	0.320	PbTe	8.4	0.050
Al ₅ Co ₂	70.0	0.276	HgNa	10.2	0.045
Mg ₂ Si	18.6	0.245	Hg ₄ Na	20.0	0.024
Li ₅ Sn ₂	66.0	0.242			

*Source: C. J. Smithells, Metals Reference Handbook, Volume II, 2nd ed., Interscience Publishers, New York, 1955.

It appears then that the number of chemical reactions which can give energy releases per unit weight of reactants equaling or exceeding those obtainable with hydrocarbon fuels are very limited. Oxidation reactions involving the very light elements and/or their hydrides appear to be the only ones capable of giving this performance. It is true that the formation of the fluorides of these elements, in general, give energy releases comparable to those obtained with oxygen and, in some cases, even somewhat better releases; however, reactions involving fluorine have not been given serious consideration here because the inconvenience associated with its use as an oxidizer is greatly outweighed by the advantage of using oxygen, particularly atmospheric oxygen. The same considerations apply to chlorine, which has a further disadvantage of giving lower energy releases in general than the reactions with oxygen. The formation of phosphides and sulfides also give lower energy releases than the oxides.

C. Fuel Processing and Regeneration Cycles

1. Abundance of Light Elements

As can be seen from the data of Table 9, most of the light elements are present in the earth's crust in sufficient quantities to provide large supplies of fuel. However, the location of deposits of an element in sufficient concentration to warrant a reasonably economic recovery is most often the limiting factor. Light elements are predominate in the first eight elements in abundance which constitute 99% of the earth's crust. These elements in decreasing order of abundance are: oxygen, silicon, aluminum, iron, calcium, sodium, potassium, and magnesium. Aluminum is the third light element in production quantity, being less only than carbon and sulfur. As shown by the data in the table, many of the other light elements are produced in significant quantities.

Table 9

ABUNDANCE AND YEARLY PRODUCTION OF LIGHT ELEMENTS*

Element	Abundance in Earth Crust (ppm)	Yearly Production and/or Consumption in the USA (tons)	Refining Process	Price (\$/lb)
Lithium	65	2500 as Li_2O (1955)	Electrolysis of fused LiCl	10
Beryllium	6	8178 as beryl ($3 \text{ BeO} \cdot \text{Al}_2\text{O}_3 \cdot 6 \text{ SiO}_2$)(1959)	Reduction of BeF with Mg -Kjellgren process	70
Boron	3	800,000 as borax ($\text{Na}_2\text{B}_4\text{O}_7 \cdot 10 \text{ H}_2\text{O}$)(1961)	Reduction of BCl_3 with H_2 in electric arc	13
Sodium	28,300	112,019 as Na metal (1959)	Castner cell - electrolysis of fused NaCl	0.17
Magnesium	20,900	30,000 as Mg metal (1959)	Electrolysis of fused MgCl_2 - Dow process	0.35
Aluminum	81,300	1.95×10^6 as Al metal (1959)	Electrochemical - Hall process	0.27
Silicon	277,200	14,000 as Si "metal" (97% Si) (1959)	Carbon reduction in electric furnace	0.19 (97% Si) 360 (semiconductor grade)
Phosphorus	1,180	367,374 as elemental P (1959)	Carbon reduction in electric furnace	0.19
Sulfur	520	5.22×10^6 as elemental S (1959)	Produced as element by Frasch process	0.025
Potassium	25,900	25 as K metal (1949)	Electrolysis in modified Castner cell	3.66
Carbon	320	810×10^6 as coal and crude oil (1958)	Mined as coal and hydrocarbons	0.05

*Source: Refs. 18, 19, and 20.

Obviously, any material used as a fuel in the transportation system would have to be made available in fairly large quantities. For example, if all the automobiles now existing in the United States were supplied with an inventory of two-hundred pounds of aluminum for fuel, the total quantity required would be around 7×10^6 tons. This is equivalent to our total aluminum production for three to four years. Once the inventory was supplied to a vehicle, replacement, except for the loss in regenerative processing, would be unnecessary. Thus the load on the primary production facilities would be reduced.

Detailed estimates of the maximum production rates of the light elements have not been made here and, indeed, such an estimate is difficult to make without knowledge of many economic factors. Therefore, the production of relatively large quantities of some light elements which are produced in only small quantities at the present time, due to limited demand, should not be discarded as a future possibility. Unless these materials have some definite advantages over those available in greater quantity and at lower cost, however, there is no incentive for their use.

2. Methods of Production Currently in Use

As shown in Table 9, the light elements are produced mainly by either of two methods, namely, electrochemical or electrolysis processes and reduction with carbon. Furthermore, even some electrochemical processes are dependent on a supply of carbon. For example, the Hall process for the production of aluminum uses consumable carbon anodes which react with the oxygen liberated from electrolysis of the aluminum oxide. Some of the elements produced in smaller quantities are prepared by reduction with magnesium. The following is a brief description of some of the more prominent processes for producing the light elements.

a. Hydrogen by Electrolysis of Water

The production of hydrogen by water electrolysis is the third most important in terms of quantity of hydrogen produced. It is exceeded by steam-hydrocarbon and steam-carbon cracking processes which both consume large quantities of carbon and hydrocarbons. Hydrogen is produced by electrolysis of a 25-35 wt-% solution of NaOH or KOH. The process is characterized by high current efficiencies (96-100%) and the cells operate at a voltage of 2.0 to 2.25 volts. Power consumption amounts to 150 kw-hr/1000 ft³ (S.T.P.) of gas. This is an energy efficiency of about 60%. The cells operate at current densities up to 200 amps/ft².

b. Sodium Production from the Castner Cell

Sodium can be produced by electrolyzing a bath of fused sodium hydroxide containing sodium chloride to improve conductivity. Water and oxygen are produced at the anode and the water so formed reacts

with one-half the sodium produced to give hydrogen. Thus, the yield of sodium can never be more than 50% of theoretical. The cells operate at a voltage of about 5 volts and current densities as high as 1800 amps/ft². The current efficiency averages 36% for long periods. Sodium is also produced by electrolysis of fused sodium chloride in the Downs process.

c. Magnesium Production by the Dow Process

Magnesium hydroxide is precipitated from sea water by the addition of lime according to the following reaction:



Magnesium chloride is formed from the hydroxide by treating with hydrochloric acid. The magnesium chloride is then added to a bath of fused chlorides from which magnesium and chlorine are formed by electrolysis. The chlorine is burned with natural gas and steam to form hydrochloric acid which is then recycled.

d. Aluminum Production by the Hall Process

Aluminum is produced by electrolysis of a bath of molten cryolite (Na_3AlF_6) containing dissolved alumina. The cell casing, lined with carbon, forms the cathode and the aluminum liberated from solution, having a slightly higher density than the electrolyte, settles to the bottom forming a layer on the carbon lining. The cell anode likewise is carbon and the oxygen released reacts with it to form carbon dioxide. In aluminum production approximately two-thirds of a pound of carbon is consumed per pound of aluminum produced and the anodes must be periodically replaced. The consumption of electrolyte is about one-tenth pound per pound of aluminum. It requires two pounds of alumina to produce one pound of aluminum. The electrical power requirement is 10 kw-hr/lb of aluminum, giving an energy efficiency of 39% (not including carbon consumed). All attempts to produce aluminum by direct carbon reduction have given poor results because of the fact that temperature required for the reduction of aluminum by carbon (1800°C) is too close to the boiling point of aluminum.

e. Silicon Production in the Electric Furnace

Silicon of 98% purity is produced by carbon reduction of pure silica in an electric furnace.

f. Phosphorus Production by Heat Electric Arc Process

Phosphorus is produced in the electric arc furnace by reaction of phosphate ores with silica and carbon. The bulk of the elemental phosphorus produced in this manner is used in the production of phosphoric acid.

A salient feature of the chemical processing cycles is that they usually involve the utilization of hydrocarbons and/or carbon for their chemical properties in the reaction in addition to their use as heat producing fuels. This is true even in a large majority of the electrolysis reactions by which light elements may be produced from their chlorides, since the regeneration of hydrochloric acid is often accomplished by burning chlorine evolved from the electrolytic cell with a hydrocarbon gas. Since nuclear energy cannot substitute for the chemical role of hydrocarbon fuels, it is necessary to devise production techniques which do not continuously consume hydrocarbons. In principle, some of the electro-refining processes which produce elements directly from their oxides, e.g., refining of aluminum, could be accomplished without using carbon. Here, it is necessary to replace consumable carbon anodes with oxidation resistant anodes. In any event, existing processes for producing light elements must be modified if nuclear energy and element ores are to be the only raw materials required. One possible modification is the use of nuclear radiation in the processing.

3. The Role of Nuclear Radiations in the Processing Cycle

The energy-producing reaction in a converter using chemical fuels is exothermic and the regeneration or processing reaction is endothermic. The promotion of chemical reactions by radiation is due to the formation of various hyperactive intermediate species such as ions, excited molecules, and free radicals. It is the formation of a greater number of these active species at a given temperature than would otherwise exist due to thermal effects alone which causes faster reaction rates. The G values (molecules of a product produced per 100 ev absorbed) for radiation initiated chemical reactions vary from 0.01 to as high as ten thousand or more. A survey of most of the G values determined to date⁽²¹⁾ shows that typical values are seldom higher than 10 with many values being 5 or less. Furthermore, endothermic reactions tend to have lower G values than exothermic reactions.

It is interesting to consider what the possible energy efficiency might be for the decomposition of light element oxides by radiation. The percentage of fission energy which finally appears in the surroundings as chemical energy in the fuel is given by $G\Delta Hx/23\%$, where,

G = G value

ΔH = heat of endothermic reaction, kcal/mol

x = fraction of fission energy released in chemical reactants
(fission recoils and nuclear radiations)

The ΔH for the decomposition of alumina is 399 kcal/mol. If we assume a G value of 5 for this endothermic reaction, and an $x = 0.2$ (attainable with oxide fuel particles 3 microns in diameter using highly enriched fuel) then

the over-all efficiency of conversion of fission energy to chemical energy as given by the above expression is $(5 \times 399 \times 0.2)/23 = 17\%$. For the electrochemical refining of aluminum, the energy efficiency is about 40%. If we assume an electric power plant conversion efficiency of 35%, then the conversion of fission to chemical energy by this path becomes $0.35 \times 0.40 = 0.14$ or 14%. This comparison points up the fact that the radiation decomposition path may be fairly attractive even though the G value is relatively low, if the ΔH of the endothermic reaction is large enough. In the case where the G value and heat of reaction are both lower, as in the radiation decomposition of water, the process is not nearly so attractive. For example, the G value for the decomposition of water is about 1, and the heat of reaction is 68 kcal/mol. With an assumed x , again, of 0.2, this gives a conversion efficiency of 0.60%. The conversion of energy by the electrolysis of water would give (for an electrolytic cell efficiency of 60%) an over-all efficiency of about 21%.

The formation of the light element oxides of interest involve ΔH 's ranging from about 150 to 400 kcal/mol. Thus, the radiation decomposition efficiencies for these fuels for an assumed $G = 5$, and an $x = 0.2$ might range from 6 to 17%. The radiation decomposition plant, at a chemical energy conversion deficiency of 17%, then has available as heat the balance of the fission energy or 83%. Presumably this energy could then be used in conventional power cycles to generate electricity. If this electrical energy is then used for electrochemical reduction, say the case of the aluminum fuel, an additional increment of $0.83 \times 14\% = 12\%$ in fuel conversion could be obtained. For the aluminum production, this would amount to an aggregate efficiency for the plant of $17\% + 12\% = 29\%$.

The greatest uncertainty in the above comparisons is that associated with the G value. While the value of 5 assumed is reasonable based on experience with a number of endothermic reactions, it would not be surprising to encounter actual values a factor of 10 lower experimentally. Reliable G values for these systems are necessary before final conclusions can be drawn. In summary, a G value of 5 for the reactions under consideration here is certainly interesting whereas a G value of 0.5 is definitely too low. Consequently, it would appear that G values greater than 2 or so would deserve closer study.

The G values for the decomposition of some solid inorganic compounds are given in Table 10. These G values, with the exception of LiNO_3 , are in the range which is interesting for the decomposition of light metal oxides in a regenerative fuel cycle. If, however, the G values for the oxides are significantly lower than the values in the table, the nuclear radiations will not play a part in the fuel regeneration step.

Table 10

YIELDS OF SOLID PHASE RADIOLYSES - A SELECTION*

(Data are for room temperature unless otherwise noted.)

Reaction	Yield	Radiation
$\text{CO}_2 \rightarrow \text{CO} + \frac{1}{2} \text{O}_2$	$G(-\text{CO}_2) = 9 \text{ } (-200^\circ\text{C})$	γ -rays
$\text{NaN}_3 \rightarrow \text{Na}^+ + 3/2 \text{N}_2 + \text{e}^-$	$G(-\text{N}_3^-) = 2 \text{ } (-200^\circ\text{C})$	X-rays
$\text{CsNO}_3 \rightarrow \text{CsNO}_2 + \frac{1}{2} \text{O}_2$	$G(\text{NO}_2^-) = 2G(\text{O}_2) = 1.7$	γ -rays, X-rays
$\text{LiNO}_3 \rightarrow \text{LiNO}_2 + \frac{1}{2} \text{O}_2$	$G(\text{NO}_3^-) = 2G(\text{O}_2) = 0.02$	γ -rays, X-rays
$\text{K ClO}_3 \rightarrow \left\{ \text{K Cl}, \text{K ClO}_2, \right.$ $\left. \text{K ClO}, \text{O}_2 \right\}$	$\left\{ \begin{array}{l} G(-\text{ClO}_3^-) = 3 \\ G(\text{O}_2) = 3 \end{array} \right.$	γ -rays, X-rays
$\text{K ClO}_4 \rightarrow 0.7 \text{ K ClO}_3$ $+ 0.3 \text{ K Cl} + 0.95 \text{ O}_2$	$\left\{ \begin{array}{l} G(-\text{ClO}_4^-) = 5 \\ G(\text{ClO}_3^-) = 3.5 \\ G(\text{Cl}^-) = 1.5 \end{array} \right.$	X-rays

*Source: Ref. 22

VI. CONCLUSIONS

This survey has been directed toward finding chemical fuels which can compete with hydrocarbon fuels on an energy storage basis. As shown, the list of materials meeting this criterion rapidly narrows to the elements below an atomic weight of 40 or so and their compounds. Furthermore, the use of atmospheric oxygen as the oxidizer for these materials has at least as good a heat of reaction as any alternate oxidizer and has the additional advantage of being available in the atmosphere. Therefore, an attractive scheme appears to be the use of atmospheric oxygen with the light elements as fuels. Of all the hydrocarbon substitutes considered, only five, namely, hydrogen, hydrogen sulfide, sulfur, ammonia, and hydrazine, give gaseous exhaust products. However, these materials and/or their combustion products all have chemical and physical properties which render them undesirable as mass quantity fuels for transportation uses. In studying the remaining materials, three stand out in terms of availability and chemical and physical properties. These elements are magnesium, aluminum, and silicon. All three approach hydrocarbon fuels in terms of energy density, and they are relatively abundant and produced in significant quantities industrially. Their chemical and physical properties make them amenable to routine handling on a large scale as surface transportation fuels. The production of all three elements would depend on the development of suitable manufacturing processes which do not consume carbon. This may be easier to accomplish in the case of aluminum since it is really only necessary that a non-consumable substitute be found for the graphite anode of the aluminum refining cell. As mentioned earlier, the processing cycle might be a dual cycle involving radiation decomposition and electrolysis, if the G values are high enough.

The type of converter which might be used with these fuels is an immediate question. Magnesium is the most reactive, and it burns readily when heated in air. Aluminum will also burn in air if it is in finely divided powder form.

Silicon is the least electropositive of the three elements and it burns with difficulty in air. Aluminum and magnesium are both very reactive metals if their protective oxide coatings are removed, whereas silicon is less reactive. Therefore, both the magnesium and aluminum can be considered for either heat engine type converters, involving combustion in air, or fuel cell converters.

Heat engines in the form of internal combustion engines and direct-cycle gas turbines are not suitable for use with fuels which form solid combustion products. There are, however, other types of engines which might be considered for use with these fuels. The Stirling, or regenerative, external combustion engine is one type, although it has not yet been adapted to small size portable power plants. Another possibility is a closed-cycle

turbine using a working fluid such as water. Thermoelectric and thermionic devices also can be readily adapted to the use of such fuels. The design of the combustion chambers for these heat engines would involve engineering problems associated with the injection of solid fuels and the removal of solid combustion products, however, it appears that many of these problems would be similar to those encountered in familiar chemical engineering unit operations such as fluidized beds and spray drying. The additional complication with these fuels is that a method for storing the solid combustion products would be required. This storage requirement is a penalty in comparison with fuels which exhaust combustion products to the atmosphere. However, since it appears to be desirable, both from the material conservation standpoint and due to the physical properties of the combustion products, this feature will be necessary if these fuels are used for large scale surface transportation applications.

In a fuel cell, the problems associated with the handling of solid reaction products are alleviated since they are dissolved in a liquid electrolyte. Fuel cells also have obvious advantages for the use of solid metallic fuels since the electrodes can be fabricated from these materials readily and easily replaced as they are consumed. The fuel cell is also easier to control and to start and stop in accordance with power needs than is the solid fuel burner. In the systems proposed here, the fuel cell would use a gaseous air electrode. To date, results with this electrode in fuel cell systems where it has been tried have been notably poor due to the dilution of the oxygen with inert nitrogen and because of limits imposed by diffusion of oxygen through electrode membranes. Means must be found for improving the performance of the air electrode in order to make fuel cells workable with atmospheric oxygen. The type of fuel cell which might evolve for a particular fuel depends on a number of factors. However, higher reaction rates give higher power densities and can be obtained with increased temperatures; therefore, the higher temperature systems using fused salt electrolytes seem to be the most probable candidates.

The important question of how best to fit nuclear energy into the future of small mobile power supplies remains unanswered. The ultimate form that the converters will take has an important bearing on this answer. It has been suggested by Weinberg that chemical fuels can be produced from carbonate rocks, water, and heat. If conventional hydrocarbon fuels can be produced in this manner, then perhaps internal combustion engines figure in the energy consumption picture for a long time to come. In any case, the use of nuclear energy to synthesize chemical fuels appears to be an important facet of a future energy distribution system. The distribution of all consumed energy as electrical power from large central station plants is not practical. Furthermore, in the future, the new types of chemical fuels that will be desired depend largely on the developments in fuel cells and batteries.

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